Effect of Nuclear Spins on Quantum Tunneling of Magnetization

In a recent Letter [1], Garg claimed that "nuclear spin dissipation can suppress MQT in magnetic particles quite severely, and cannot in any case be neglected for a quantitative understanding of the problem." The purpose of this Comment is to demonstrate that the effect of nuclear spins on magnetic tunneling is far less dramatic than suggested in Ref. [1]; in most systems it can be ignored, at least at the present stage of the experiment. Given crude approximations of the Letter, it can only pretend (as well as this Comment) for a rough estimate of the effect. In this case no calculation is needed, the answer can be obtained in terms of characteristic fields commonly used in NMR studies. The electronic, $\mathbf{M}$, and nuclear, $\mathbf{m}$, magnetizations are coupled via the hyperfine interaction, $E_{\rm int} = A\mathbf{M}\cdot\mathbf{m} = -\mathbf{H}_{\rm hf}\cdot\mathbf{m} = -\mathbf{h}_{\rm hf}\cdot\mathbf{M}$ (A being a constant). Here $\mathbf{H}_{\rm hf} = -A\mathbf{M}$ is the hyperfine field on the nucleus, while $\mathbf{h}_{\rm hf} = -A\mathbf{m}$ is the hyperfine field exerted by the nuclear moments on the electronic magnetization. $\mathbf{M}$ satisfies $\dot{\mathbf{M}} = \gamma\mathbf{M}\times\mathbf{H}_{\rm eff}$, where $\mathbf{H}_{\rm eff} = -\delta E/\delta\mathbf{M}$ (E being the total magnetic energy). In the model [2] studied by Garg, the barrier for the tunneling is lowered by tunneling the external field H to the anisotropy field H_a. In that model $H_{\rm eff} = \epsilon H_a + h_{\rm hf}$, where $\epsilon = 1 - H/H_a$ characterizes the height of the barrier, $U \propto \epsilon^2$. Consequently, $h_{\rm hf}/H_a\epsilon$ determines the relative perturbation of the real- and imaginary-time dynamics of $\mathbf{M}$ by nuclear spins. The corresponding perturbation of the tunneling exponent is $B = B_0(1 + h_{\rm hf}/H_a\epsilon)$. This is exactly what one finds from Eq. (21) of Ref. [1] if its parameters μ and η are expressed in terms of the relevant fields and the physically relevant limit (see below) of $\mu\eta \ll 1$ is applied. The tunneling exponent is proportional to the volume of a single domain particle and must be kept around 30 to insure observation of tunneling on the time scale of the experiment. For that reason a "severe suppression" of tunneling by $h_{\rm hf}/H_a\epsilon{\sim}0.3$, claimed by Garg, is an overstatement, as it would only require a 10% decrease in the size of the particles to restore the desired value of the tunneling rate. However, even such a value of $h_{\rm hf}/H_a\epsilon$ should be viewed as rather high for most systems. It is obvious (as in other MQT problems) that in the limit of $\epsilon \to 0$ when the barrier becomes small, the contribution of nuclear spins, as well as of other unaccounted interactions, may, in principle, become important. However, very small ϵ should not be realistic; for the nuclear spin dissipation to be notable one needs a not very small ratio $h_{\rm hf}/H_a$. The nuclear magnetization, to which $h_{\rm hf}$ is proportional, satisfies the Curie-Weiss law, so that the largest effect occurs at $k_BT \ll \mu_n H_{\rm hf}$ (μ_n being the moment of the nucleus). Table I summarizes relevant isotopes, their natural abundance, nuclear moments, nuclear ordering temperature, as well as the zero temperature values of the nuclear magnetization m, of the field $h_{\rm hf}$, and of the expected ratio $h_{\rm hf}/H_a$ for five commonly used ferromagnetic elements. The anisotropy field responsible for the barrier has been taken in the kOe range for Fe and Ni, and in the range of tens of kOe for Co, Tb, and Dy, which should be a reasonable estimate for particles of size less than 100 Å. As follows from Table I, the interaction with nuclear spins should be unimportant in Fe- and Ni-based ferromagnetic compounds. It must also be irrelevant in antiferromagnets [3,4] to which $\epsilon\to 0$ is not applied. Below ordering temperature, nuclear spins might, in principle, play some role in Co and Tb which have large nuclear moments and 100% concentration of the isotope. However, even in this case, one needs $\epsilon{\sim}10^{-2}$ to compensate the smallness of $h_{\rm hf}/H_a$.

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TABLE I. Nuclear magnetism in fine particles of five commonly used ferromagnetic materials.

Material	Isotope	Abundance (%)	μ_n (nm)	$\mu_n H_{\rm hf}$ (mK)	m (emu/cm^3)	$h_{\rm hf}$ (Oe)	$h_{\rm hf}/H_a$
Iron	^{57}Fe	2.2	0.090	1.1	8.1×10^{-4}	0.16	$\sim 10^{-4}$
Cobalt	^{59}Co	100	4.6	38	2.1	330	$\sim 10^{-2}$
Nickel	^{61}Ni	1.2	0.75	2.0	4.0×10^{-3}	0.59	$\sim 10^{-4}$
Terbium	^{159}Tb	100	2.0	220	0.31	360	$\sim 10^{-2}$
Dysprosium	$\frac{^{161}\text{Dy}}{^{163}\text{Dy}}$	$\frac{19}{25}$	$\frac{0.46}{0.64}$	$\frac{99}{130}$	$\frac{1.4}{2.6}\times10^{-2}$	$\frac{27}{49}$	$\sim 10^{-3}$

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